TEMPERATURE DEPENDENCE OF THE OPTICAL ABSORPTION BANDWIDTH OF MnF_2 *CRYSTALS*

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A strong temperature dependence of the optical absorption bandwidth was found for MnF_2 crystals in the temperature range below the Néel point. The bandwidth in the paramagnetic region depended weakly on temperature. In this connection it is suggested that the profile and the width of the optical absorption bands of the antiferromagnetic MnF_2 crystals are governed not by the usual interaction with phonons but by interaction with oscillations of the ionic magnetic moments.

In earlier investigations of the effect of antiferromagnetic ordering on the optical absorption spectra of MnF_2 crystals,[1,2] attention has mainly been paid to the temperature shift of the bands along the frequency scale. No clear anomaly of the temperature dependence of the band shift was found near the Néel temperature ($T = 68°K$). It seemed to us more interesting to investigate the temperature dependence of the optical absorption bandwidth.

Of all the bands observed in the absorption spectrum of MnF_2 crystals, the C-group (Stout's notation[1]) is the most convenient for this purpose: it is a doublet of bands which are sufficiently strong in a wide range of temperatures. Isolated values of the half-widths of the C-bands, taken from tables or estimated from published spectrograms, are collected in Fig. 1. Here triangles denote the results obtained in unpolarized light;[2] they are of no value to us. The black dots represent the σ-component (light polarized at right angles to a four-fold axis, i.e., with the electric field $\mathbf{E} \perp \mathbf{c}$) and open circles represent the π-component ($\mathbf{E} \parallel \mathbf{c}$). These results show a marked kink in the temperature dependence of the half-width. However, we must remember that these results may be strongly distorted, since Stout was interested mainly in the temperature shift of the bands and estimated the half-widths from spectrograms without any allowance for the large changes in the band intensity on cooling.

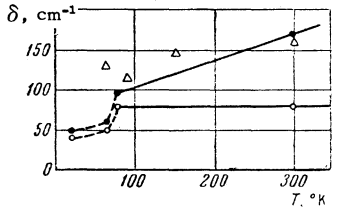

FIG. 1. Temperature dependence of the C-band half-widths: Δ – results of [2]; ●, o – results of [1].

Because of this we measured carefully the variation of the profile and intensity of the π- and σ-components of the C-bands in a wide range of temperatures: at room temperature (300°K), at an intermediate temperature ($\approx$ 180°K), at the boiling points of liquid oxygen, nitrogen and hydrogen (90, 77 and 20°K), and, finally, at temperatures of 65 and 55°K obtained by pumping nitrogen and oxygen respectively.

Measurements were carried out in polarized light using the high-dispersion spectrographs DFS-8 (6 Å/mm) and DFS-3 (4 Å/mm). The absorption intensity was measured by the usual photographic photometry method with a multistep attenuator.[3]

Figure 2 shows examples of the absorption-coefficient (K) spectra in the C-band region for three temperatures. From such curves we determined the band half-widths and plotted their temperature dependences (Fig. 3).

Figure 3 illustrates the prominent singularity in the temperature dependence of the half-width of the π- and σ-components of the C-band at the antiferromagnetic ordering point ($T = 68°K$). The temperature dependence of the D-band half-width (λ = 3567.85 A at 20°K), denoted by triangles in Fig. 3, is obviously similar.

As far as we know this is the first observation of this type of temperature dependence of the optical absorption bandwidths. Usually the optical absorption bandwidths are governed by the interaction of electron transitions with lattice vibrations. Thus Toyozawa[4,5] showed that the width of the exciton absorption bands is determined by

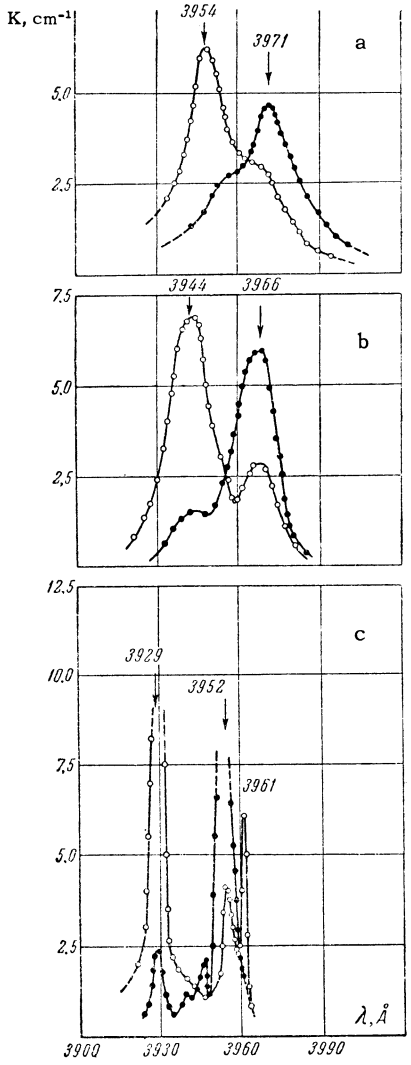

FIG. 2. Absorption coefficient spectra of MnF_2 in the C-band region at temperatures of : a) 300° K; b) 90° K; c) 20° K; ●–E ⊥ c, ○–E ∥ c.

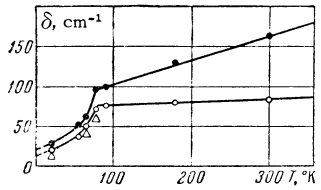

FIG. 3. Temperature dependence of the half-widths of the C and D absorption bands: ●– C-band, E ⊥ c (σ-component); ○– C-band, E ∥ c (π-component); △– D-band (λ = 3567.9 A), E ⊥ c.

the exciton-phonon interaction and the temperature dependence of the bandwidth at not too low temperatures is linear for weak exciton-phonon coupling. This behavior is observed experimentally in the case of the optical absorption by MnF_2 single crystals at temperatures above T = 68°K.

It is interesting to note that Toyozawa also explained the asymmetry of the exciton absorption bands. From the standpoint of his theory the "shading" of the σ-component at long wavelengths (at 20°K, λ = 3952 Å and the asymmetry is ≈ 30%) may indicate that the effective mass in the corresponding exciton band is negative near the zero value of the quasimomentum (k = 0).

It is quite clear that the observed singularity in the temperature dependence of the absorption bandwidth is related to the antiferromagnetic ordering. It is not very likely that at the Néel point, T_N, the phonon spectrum or the intensity of the exciton-phonon interaction change greatly, since the crystal lattice parameters alter only slightly.[6] Therefore, although there is no rigorous theory, it seems likely that the profile and width of the optical absorption bands of antiferromagnetic crystals below $T = T_N$ are governed by an interaction with excitations of spin-wave type and not with phonons.

The observed temperature dependence of the half-width can be understood as follows. At T = 0°K the magnetic structure is fully ordered and the optical absorption bands are quite narrow. Increase of the temperature disturbs the magnetic order and strongly broadens the absorption bands, whose width rapidly rises with increasing temperature up to $T = T_N$, where the magnetic structure is fully disordered and the phonon mechanism of band broadening begins to play the dominant role.

A complete explanation of the observed singularity in the temperature dependence of the absorption bandwidth will only be given by a rigorous theory.

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